FORM PTO-1390 (REV 11-98) U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY DOCKET NUMBER: 2119-4203

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

U S APPLICATION NO (If known, see 37 CFR 1 51): 09/600621

INTERNATIONAL APPLICATION	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP98/08480	28 December 1998	20 January 1998

TITLE OF INVENTION
LOW PIGMENT CONTENT POLYESTER FILM AND PROCESS FOR MAKING THE SAME

APPLICANT(S) FOR DO/EO/US
Christine ANDREIS; Gusty FEYDER

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is **FIRST** submission of items concerning a filing under 35 U S C. 371
2. ☐ This is **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U S. C 371 (b) and PCT Articles 22 and 39 (1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International application into English (35 U S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U S C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C 371(c)(4)) _____
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5))

Items 11. to 16. below concern document(s) or information included.

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1 98
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included
13. ☒ A FIRST preliminary amendment
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☐ Other items or Information:

U S APPLICATION NO (if known, see 37 C F R 1 51)	INTERNATIONAL APPLICATION NO	ATTORNEY'S DOCKET NO
Unassigned 09/600621	PCT/EP98/08480	2119-4203

17. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):	CALCULATIONS PTO USE ONLY
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2) paid to USPTO and International Search Report not prepared by the EPO or JPO $970 00	
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO $840.00	
International preliminary examination fee (37 CFR 1 482) not paid to USPTO but international search fee (37 CFR 1 445(a)(2) paid to USPTO $760.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33 (1) - (4) $670.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1) - (4) $96.00	
ENTER APPROPRIATE BASIC FEE AMOUNT =	$840.00
Surcharge of **$130** for furnishing the oath or declaration later than ☐20 ☒30 months from the earliest claimed priority date (37 CFR 1 492(e)).	$130.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	26 - 20 =	6	X $18.00	$108.00
Independent claims	1 - 3 =	0	X $78.00	$ 0.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ $260 00	$

TOTAL OF ABOVE CALCULATIONS =	$1,078.00
Reduction of ½ for filing by small entity, if applicable. A Small Entity Statement must also by filed (Note 37 CFR 1.9, 1.27, 1.28).	$
SUBTOTAL =	$1,078.00
Processing fee of **$130.00** for furnishing the English translation later than ☐20 ☐30 months from the earliest claimed priority date (37 CFR 1.492(f)). +	$
TOTAL NATIONAL FEE =	**$1,078.00**
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3 31). **$40.00** per property +	$
TOTAL FEES ENCLOSED	$1,078.00
Amount to be refunded:	$
charged	$1,078.00

a. ☐ A check in the amount of $ to cover the above fees is enclosed.

b ☒ Please charge my Deposit Account No. 13-4500 in the amount of $ 1,078.00 to cover the above fees.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-4500. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Morgan & Finnegan, L.L.P.
345 Park Avenue
New York, New York 10154
(212) 758-4800 Telephone
(212) 751-6849 Facsimile

SIGNATURE
Michael S. MARCUS
NAME
31,727 July 20, 2000
REGISTRATION NO. DATE

15770_1

PATENT Our Docket No. 2119-4203

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Christine ANDREIS
Gusty FEYDER

Serial No.: Unassigned Group Art Unit: Unassigned

Filed: July 20, 2000

For: LOW PIGMENT CONTENT POLYESTER FILM AND PROCESS FOR MAKING THE SAME

Commissioner of Patents
U.S. Patent & Trademark Office
Washington, DC 20231

PRELIMINARY AMENDMENT

Sir:

Kindly amend the above-identified application prior to examination on the merits as follows:

IN THE CLAIMS:

3. (Amended) The film [of] according to claim 1 [or 2] where the ratio is [comprised] between 1 and 10[, preferably 2 and 5].

4. (Amended) The film [of any one of] according to claim[s] 1 [to 3], wherein the at least one layer[(s)] containing a pigment [contain(s)] contains more than 10 wt% of pigment[, preferably more than 15 wt% of pigment, most preferably more than 20 wt% of pigment].

5. (Amended) The film [of any one of] according to claim[s] 1[to 4], wherein the at least one layer[(s)] containing a pigment [contain(s)] contains less than 50 wt% of pigment[, preferably less than 40 wt% of pigment].

6. (Amended) The film [of any one of] according to claim[s] 1 [to 5], wherein each layer has a thickness [comprised] in a range of between 1μm and 200μm[, preferably between 2 and 50μm].

7. (Amended) The film [of any one of] according to claim[s] 1 [to 6], wherein the pigment has a particle size in a range of between 0.01μm and 5μm[, preferably between 0.02 and 1.0μm].

8. (Amended) The film [of any one of] according to claim[s] 1 [to 7], wherein the layers are coextruded layers.

9. (Amended) The film [of any one of] according to claim[s]1 [to 8], wherein the pigment is titanium dioxide.

10. (Amended) The film [of any one of] according to claim[s] 1 [to 8], wherein the polyester is PET.

11. (Amended) A process for making the film of [any one of] claim[s] 1 [to 10], comprising the step of coextruding [the various] said layers.

Please add the following new claims:

--13. The film according to claim 3 where the ratio is between 2 and 5.

14. The film according to claim 2 where the ratio is between 1 and 10.

15. The film according to claim 14 wherein the ratio is between 2 and 5.

16. The film according to claim 4, wherein the pigment is present in an amount of greater than 15 wt%.

17. The film according to claim 16, wherein the pigment is present in an amount of greater than 20 wt%.

18. The film according to claim 5, wherein the at least one layer containing a pigment contains less than 40 wt% of pigment.

19. The film according to claim 6, wherein each layer has a thickness in the range of between 2μm and 50μm.

20. The film according to claim 7, wherein the pigment has a particle size in the range of between 0.02μm and 1.0μm.

21. The film according to claim 2, wherein the layers are coextruded layers.

22. The film according to claim 2, wherein the pigment is titanium dioxide.

23. The film according to claim 2, wherein the polyester is PET.

24. A process for making the film of claim 2, comprising the step of coextruding said layers.

25. A process for making the film of claim 3, comprising the step of coextruding said layers.

26. A process for making the film of claim 14, comprising the step of coextruding said layers.

REMARKS

Original claims 1-12 according to the amended sheets and newly added Claims 13-25 are pending in the application. Newly added claims 13-25 are fully supported by the original specification and claims. No new matter has been introduced. Examination on the merits is respectfully requested.

AUTHORIZATIONS:.

The Commissioner is hereby authorized to charge any additional fees which may be required for the consideration of this preliminary amendment pursuant to 37 CFR §§ 1.16 and 1.17, or credit any overpayment to Deposit Account No. 13-4500, Order No. 2119-4203.

Respectfully submitted,

MORGAN & FINNEGAN, L.L.P.

Dated: July 20, 2000

By: ____________________
Michael S. Marcus
Registration No. 31,727
(202) 857-7887 Telephone
(202) 857-7929 Facsimile

CORRESPONDENCE ADDRESS:
MORGAN & FINNEGAN, L.L.P.
345 Park Avenue
New York, New York 10154
(212) 758-4800 Facsimile
(212) 751-6849 Telecopier

LOW PIGMENT CONTENT POLYESTER FILM AND PROCESS FOR MAKING THE SAME

TECHNICAL BACKGROUND OF THE INVENTION

The instant invention relates to a low pigment content polyester film, as well as to a process for making the same.

Pigments, especially white pigments, are known additives widely used to impart opacity or specific light transmission properties to films. However, there is currently no film designed for the sole purpose of containing (white) pigments, although there is a need for such films in applications where controlled haze, light transmission, opacity, etc., are desired final properties.

A film comprised of a mere layer containing pigment is not satisfactory, since the incorporation of fillers, especially pigments, although imparting valuable light transmission properties, presents the drawback of lowering the mechanical properties of the final film, compared to those of the host polymer prior to incorporation of the filler.

Films are widely known, and afford to combine various properties of different layers that are associated together. For example, films based on polyester are known.

JP-A-8290539 (to Diafoil Hoechst Co) discloses polyester films where the inner layer comprises fluorescent fine particles and the outer layer comprises white pigment. Said film is said to be adapted as a magnetic recording medium. The amount of white pigment is however very low.

Similarly, JP-A-8045067 (to Diafoil Hoechst Co) discloses polyester films where the inner layer comprises reclaimed raw material and the outer layer comprises white

pigment. Said film is said to be adapted as a magnetic card on a pin ball game. The amount of white pigment is however very low.

JP-A-4110147 and JP-A-4110148 (to Diafoil Hoechst Co) disclose polyester films where one of the layers comprises a micropowder in an amount of from 0.1 to 5 wt%. The film has a highly delustered surface and is used for transfer moulding.

JP-A-6000859 (to Diafoil Hoechst Co) discloses polyester films where one of the layers comprises a micropowder in an amount of from 0.001 to 20 wt%, especially from 0.01 to 10 wt%. The film is said to be adapted as a magnetic recording medium, as well as a packing film, a matted film, etc.. All examples make however use of very low amounts of the filler, typically about 1 wt%. Further, the process specified in said patent application is based (i) on extrusion-lamination; (ii) direct feeding with the result of the continuous polymerisation process and (iii) mixing of the filler or additive at the extruder level. This process does not make use of "chips" of polyester, and the feeding of the filler or additive at the extruder level makes it almost impossible to obtain a uniform dispersion of the filler or additive for high amounts, e.g. above 5 wt%.

Although films where one layer contains pigments are known from the above publications, it should be emphasized that these films have not been designed for the sole purposes of containing a white pigment. The problem faced when high amounts of pigments required for imparting high opacity are incorporated is that the mechanical properties of the final film are reduced. It is in fact well established that high opacity and high mechanical properties cannot be found in the same film. Further, it is also well established that high opacity requires high pigment content. Thus there is a need for a film that would show good mechanical properties together with good opacity properties, while minimizing the overall pigment content.

SUMMARY OF THE INVENTION

The invention thus provides such a film, in the form of a film comprising:

(i) at least one polyester layer containing more than 5 wt% of a pigment, and

(ii) at least one polyester layer substantially devoid of pigment,

where the ratio of the respective thickness of the layer(s) devoid of pigment to the pigmented layer(s) is at least 1.

The invention allows to combine the light transmission properties of a layer having a high pigment content together with the traditional mechanical properties of a layer substantially devoid of pigments.

DESCRIPTION OF EMBODIMENTS OF THE INVENTION.

The polyester used in the invention is any polyester where the major part of it is comprised of any aromatic repeating ester units. The term polyester in this invention refers to a polymer that is obtained by condensation polymerization of an aromatic dicarboxylic acid such as terephthalic acid or 2,6-naphthalene dicarboxylic acid and of an aliphatic terephthalic acid glycol such as ethylene glycol, 1,4-butanediol or 1,4-cyclohexane dimethanol. These polymers, in addition to being homopolymers, may also be copolymers having a third component or serveral components. In this case, the dicarboxylic acid component may be, for example, isophthalic acid, phthalic acid, terephthalic acid, 2,6-naphthalene dicarboxylic acid, 4,4'-diphenyldicarboxylic acid, adipic acid, sebacic acid, decanedicarboxylic acid and 1,4-cyclohexane dicarboyxlic acid; the oxycarboxylic acid component can be, for example, p-oxybenzoic acid and the glycol component can be, for example, ethylene glycol, diethylene glycol, triethylene glycol, propylene glycol, butanediol, neopentyl glycol, 1,4-cyclohexane dimethanol, polyethylene glycol and polytetramethylene glycol. Examples of such polyesters are polyethylenenaphthalate (PEN), polybutyleneterephthalate (PBT), polyethyleneterephthalate (PET), the latter being the preferred polyester. Mixtures are also possible,

optionally with another polymer different from a polyester. The intrinsic viscosity of the polyester that is used in the invention may vary from e.g. 0.45 to e.g. 0.7, measured in phenoltetrachloreethane at 30°C. The MW may vary within broad limits, e.g. between 10000 to 30000 g/mol.

Examples of pigments are TiO_2, SiO_2, $CaCO_3$, Al_2O_3, $BaSO_4$, carbon black, zeolite, kaolin, the preferred one being TiO_2. Mixtures are also possible. Preferably, the pigment is a white pigment. Voids can also be provided around the particles of pigment, as taught by EP-A-0688814. The pigment used can be the same or different in the pigmented layers, when more than one is used. The pigment has a particle size usually comprised between 0.01 and 5μm, for example between 0.02 and 1.0μm.

The following description is given with reference to PET and TiO_2 for the sake of convenience only; it shall not be construed as limited to these embodiments.

The layer containing the pigment will contain at least 5 wt%, preferably at least 10 wt%, especially at least 15 wt% of pigment, most preferably more than 20 wt% of pigment, while the layer substantially devoid of pigment will contain less than 1wt%, and is preferably without pigment.

Upper limits for the content of pigment can be less than 50 wt% of pigment, preferably less than 40 wt% of pigment.

Thus, ranges for the pigment content are, e.g. 15-50 wt%, preferably 20-40 wt%.

Conventional additives may be incorporated in the layers of the instant films, in conventional amounts. Examples thereof are anti-oxidant, anti-UV agents, etc.. Mixtures are also possible.

The instant film formed of the various layers can be of various structures and the polyester used can vary from one layer to another. For example, the inner pigment-free layer and/or the outer pigmented layer(s) can be obtained from starting products containing scrap material, allowing recycling. For example, the inner layer can contain scrap

material, while the pigmented layers do not. The inner layer may even contain scrap material of the final film of the invention, provided the content of pigment is less than 1 wt%. Content of scrap is variable within broad limits known to the skilled man. One pigmented layer can be mat while the other is glossy. Also, the polyester can have a different nature from one layer to another, or they can be the same. All layers can be comprised of crystalline polyester, or all layer can be comprised of amorphous copolyester, or one layer can be crystalline and the other(s) can be amorphous. Especially, the invention provides films where the inner layer is crystalline (i.e. PET) while one or two of the outer layer(s) is amorphous (i.e. copolyester). This allows to combine specific mechanical properties of crystalline polyester and adhesive properties of the copolyester. It can also be forseen that each layer may be formed of two or more sub-layers; e.g. the outer pigmented layer can be formed of one sub-layer of PET and one sub-layer of copoyester, the first one being in contact with the inner layer.

The instant films can be used as individual layers in further multi-layers films. When necessary, a binder known to the skilled man can be used to incorporate the instant films into a further multi-layers arrangement.

The thickness of each layer can vary within broad limits, for example between 1 and 200µm, preferably between 2 and 50µm. One specific embodiment is the film where the layer substantially devoid of pigment represents the major part of the PET thickness; especially the ratio [thickness PET]/[thickness PET+TiO_2] can be comprised between 1 and 10, preferably 2 and 5.

The invention also provides a process for making the instant films, where the layers are coextruded. Coextrusion operative conditions are within the general knowledge of the skilled man. Other processes such as lamination, calendering, etc. can also be contemplated.

One preferred process makes use of a master-batch, where the pigment represents, e.g. 50wt%. The pellets or

chips of said master-batch are then mixed with pure polyester pellets or chips and fed into the extruder. The master-batch can also be mixed with chips of recycled (coextruded) films, thus possibly already containing a small amount of pigment. This allows to have a very uniform dispersion of the pigment in the polyester, which would otherwise not be obtainable by direct feeding of the pigment into the extruder.

The following examples are provided for illustrative purposes only and shall not be considered as limiting the scope of the invention.

COMPARATIVE EXAMPLES

Comparative Example 1.

A resin composition consisting of 95 wt% of PET and 5 wt% of anataze-type titianium oxide was melt extruded at 280°C by a twin-screw extruder through a T shaped die and cooled down on a chill-roll, which was water-cooled at 30°C. Then, the non-drawn sheet was stretched at 80°C by a roll stretching machine at a draw ratio of 3.4 in the MD and then at 130°C by a tenter at a draw ratio of 3.8 in the TD, and fixed at 230°C by heat while relaxing the sheet at a rate of 4%. The resulting film was 20μm thick with the average particle size of the fine titanium oxide particles being less than 0.5μm.

Comparative Example 2.

A 20μm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 92.5 wt% of PET and 7.5 wt% of anataze-type tinanium oxide is used.

Comparative Example 3.

A 20μm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 90 wt% of PET and 10 wt% of anataze-type tinanium oxide is used.

Comparative Example 4.

A 20μm film was obtained by the same procedure as in comparative example 1 except that a resin composition

consisting of 86 wt% of PET and 14 wt% of anataze-type tinanium oxide is used.

Comparative Example 5.

A 40µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 95 wt% of PET and 5 wt% of anataze-type tinanium oxide is used.

Comparative Example 6.

A 40µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 91 wt% of PET and 9 wt% of anataze-type tinanium oxide is used.

Comparative Example 7.

A 12µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 95 wt% of PET and 5 wt% of anataze-type tinanium oxide is used.

Comparative Example 8.

A 12µm film was obtained by the same procedure as in comparative example 1 except that a resin composition consisting of 90 wt% of PET and 10 wt% of anataze-type tinanium oxide is used.

WORKING EXAMPLES.

Example 1.

A resin I composition consisting of 95 wt% of PET and 5 wt% of anataze-type titianium oxide and a resin II composition consisting of 100 wt% of PET are melt extruded at 280°C in two twin-screw extruders through a coestrusion-feedblock in such a way that resin I is separated in two melt streams to form a sandwich structure with resin II as the inner layer. The three layers cast is cooled down on a chill-roll, which was water-cooled at 30°C. Then, the non-drawn 3 layers cast is stretched as in the comparative examples. The resulting film is 15µm thick with a thickness distribution [PET/TiO_2]/PET[PET/TiO_2] of 3.5/8/3.5. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 1.14 and the overall TiO_2 content is the 3 layers film is 2.33 wt%.

Example 2

A 15µm coextruded white film was mobtained by the same procedure as in example 1 except that the composition of resin I was 93% by weight of polyethylene terephthalate and 7% by weight anataze-type TiO_2. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 1.14 and the overall TiO_2 content in the 3 layer film is 3.27 wt%.

Example 3

A 15µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 91% by weight of polyethylene terephthalate and 9% by weight anataze-type TiO_2. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 1.14 and the overall TiO_2 content in the 3 layer film is 4.2 wt%.

Example 4

A 15µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 89% by weight of polyethylene terephthalate and 11% by weight anataze-type TiO_2. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 1.14 and the overall TiO_2 content in the 3 layer film is 5.13 wt%.

Example 5

A 15µm coextruded white film was obtained by the same procedure as in Exazmple 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anataze-type TiO_2. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 1.14 and the overall TiO_2 content in the 3 layer film is 12.25 wt%.

Example 6

A 16µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 81% by weight of polyethylene terephthalate and 19% by weight anataze-type TiO_2. The thickness ratio [PET-TiO_2]/PET/[PET-TiO_2] was 2µM/12µm/2µm. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 3 and the overall TiO_2 content in the 3 layer film is 4.75 wt%.

Example 7

A 15µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 74% by weight of polyethylene terephthalate and 26% by weight anataze-type TiO_2. The thickness ratio [PET-TiO_2]/PET/[PET-TiO_2] was 2µm/11µm/2µm. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 2.75 and the overall TiO_2 content in the 3 layer film is 6.93 wt%.

Example 8

A 20µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 75% by weight of polyethylene terephthalate and 25% by weight anataze-type TiO_2. The thickness ratio [PET-TiO_2]/PET/[PET-TiO_2] was 2µm/16µm/2µm. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 4 and the overall TiO_2 content in the 3 layer film is 5 wt%.

Example 9

A 20µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anataze-type TiO_2. The thickness ratio [PET-TiO_2]/PET/[PET-TiO_2] was 3.5µm/13µm/3.5µm. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 1.86 and the overall TiO_2 content in the 3 layer film is 4.9wt%.

Example 10

A 40µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anataze-type TiO_2. The thickness ratio [PET-TiO_2]/PET/[PET-TiO_2] was 3.5µm/33µm/3.5µm. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 4.71 and the overall TiO_2 content in the 3 layer film is 2.45 wt%.

Example 11

A 40µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 85% by weight of polyethylene terephthalate and 15% by weight anataze-type TiO_2. The thickness ratio [PET-TiO_2]/PET/[PET-TiO_2] was 5µm/30µm/5µm. The overall ratio

[thickness PET]/[thickness PET+TiO_2] is 3 and the overall TiO_2 content in the 3 layer film is 3.75 wt%.

Example 12

A 20µm coextruded white film was obtained by the same procedure as in Example 1 except that the composition of resin I was 86% by weight of polyethylene terephthalate and 14% by weight anataze-type TiO_2. The thickness ratio [PET-TiO_2]/PET/[PET-TiO_2] was 5µm/10µm/5µm. The overall ratio [thickness PET]/[thickness PET+TiO_2] is 1 and the overall TiO_2 content in the 3 layer film is 7wt%.

The following properties were measured on some films of comparative examples and some films of working examples.

(1) Opacity

Opacity is the hiding power of a non-transparent material. It was measured with a Hunterlab Colorquest spectrophotometer in reflection mode using illuminant III VC (wavelength 400 to 680nm). A black and white reference was taken to determine the opacity given in percent. A completely transparent film is equal to 0% opacity.

(2) Whiteness index

Whiteness index is associated with a region or volume in the color space in which objects are recognized as white. The degree of whiteness is measured by the degree of departure from a perfect white (ideal white is bluish). It was measured with a Hunterlab Colorquest spectrophotometer in transmission mode using illuminant III C.

(3) Isothermal shrinkage

The size of a 5x5 inch sample was measured before and after heat treatment in an oven at different temperatures, namely 150°C and 200°C. The difference in size is measured and expressed in %, expressing stability.

(4) Mechanical properties

Modulus, tensile strengh, force at 3% elongation (F3), force at 5% elongation (F5) and elongation at break were determined with an Instron equipment at room temperature.

Tables 1 and 2 give the data for the comparative examples and the working examples, respectively, where Th is thickness, Thcoex is thickness of pigmented layer,

CoexTiO$_2$% is the % of TiO$_2$ in the pigmented layer, total TiO$_2$% is the % of TiO$_2$ in the film. Table 3 gives mechanical data for films of comparative examples 1 and 2 and working examples 7 and 8.

Table 1

Comp. Ex.	Th (µm)	TiO2(%)	Opacity %	Whiteness index	DS 150(%)	DS 200(%)
1	20	5	57.70	-	2.2	8.2
2	20	7.5	64.50	27.3	2.8	9.2
3	20	10	71.7	16.5	2.8	9.2
4	20	14	82.2	-	2.4	8.4
5	40	5	66.7	-	3.2	10.2
6	40	9	81.5	-	3.0	9.4
7	12	5	49.6	-	-	
8	12	10	66.0	-	3.6	10.6

Table 2

Example	Th (µm)	Thcoex (µm)	Coex TiO2%	Total TiO2%	Opacity %	Whiteness index	DS 150(%)	DS 200(%)
1	15	3.5	5	2.3	39.30	-	2.0	8.0
2	15	3.5	7	3.3	47.7	-	2.0	7.2
3	15	3.5	9	4.2	51.4	-	1.8	6.8
4	15	3.5	11	5.1	56.8	-	1.8	6.6
5	15	3.5	14	12.3	61.6	-	1.6	6.2
6	16	3.5	19	4.8	72.9	20.0	1.6	6.0
7	15	2	26	6.9	65.0	-	1.8	6.6
8	20	2	25	5	66.10	-	1.8	6.8
9	20	3.5	14	4.9	64.0	29.8	1.4	6.0
10	40	3.5	14	2.5	61.8	-	2.6	8.4
11	40	5	15	3.8	66.9	-	2.2	8.8
12	20	5	14	7	69.0	-	1.2	6.6

Table 3

	Ex. 7	Comp. ex. 2	Ex. 8	Comp. ex. 1
Thickness (µm)	15	20	20	20
TiO_2(%)	6,9	7,5	5	5
total	26	7.5	25	5
coex				
Opacity	65	64	66	57
Modulus MD	3669	3865	4370	4069
(Mpa) TD	4992	4580	4832	4325
F3 MD	84	87	90	88
(N/mm^2) TD	93	93	94	89
F5 MD	99	101	103	105
(N/mm^2) TD	103	104	103	99
Tensile MD	184	201	236	215
(N/mm^2) TD	258	224	262	221
Elong. MD	138	113	117	111
(N/mm^2) TD	259	224	262	221

By comparing tables 1 and 2, it is possible to conclude the following:

for a given total Tio_2 content and thickness, opacity is higher for the coextruded 3 layer films than for the completely filled monolayer films.

for a given opacity, the whiteness index is higher for the films of the invention than for the completely filled monolayer films, evidencing that the coextruded films need less pigment.

for a given opacity, the isothermal shrinkage of the coextruded films is lower than that of the monolayer film, evidencing that the films of the invention are more stable.

By analyzing the content of table 3, it is possible to conclude that all mechanical values obtained for the coextruded 3 layers films are higher than those obtained for the monolayer film, evidencing superiority of the instant films.

The present invention is not limited by the embodiments described above but shall be construed in accordance with the appended claims.

CLAIMS.

1. A film comprising:
(i) at least one crystalline polyester layer containing more than 5 wt% of a pigment, and
(ii) at least one crystalline polyester layer substantially devoid of pigment,
where the ratio of the respective thickness of the layer(s) devoid of pigment to the pigmented layer(s) is at least 1.

2. The film of claim 1 comprising two outer layers containing pigment and one inner layer substantially devoid of pigment.

3. The film of claim 1 or 2 where the ratio is comprised between 1 and 10, preferably 2 and 5.

4. The film of any one of claims 1 to 3, wherein the layer(s) containing a pigment contain(s) more than 10 wt% of pigment, preferably more than 15 wt% of pigment, most preferably more than 20 wt% of pigment.

5. The film of any one of claims 1 to 4, wherein the layer(s) containing a pigment contain(s) less than 50 wt% of pigment, preferably less than 40 wt% of pigment.

6. The film of any one of claims 1 to 5, wherein each layer has a thickness comprised between 1 and 200μm, preferably between 2 and 50μm.

7. The film of any one of claims 1 to 6, wherein the pigment has a particle size comprised between 0.01 and 5μm, preferably between 0.02 and 1.0μm.

8. The film of any one of claims 1 to 7, wherein the layers are coextruded layers.

9. The film of any one of claims 1 to 8, wherein the pigment is titanium dioxide.

10. The film of any one of claims 1 to 8, wherein the polyester is PET.

11. A process for making the film of any one of claims 1 to 10, comprising the step of coextruding the various layers.

12. The process of claim 11, comprising the step of using a master-batch.

Rev. 06/95

Docket Number
IM-1175 US NA

DECLARATION and POWER OF ATTORNEY

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Low Pigment Content Polyester Film and Process of Making the Same

the specification of which is attached hereto unless the following box is checked:

■ was filed on **20 July 2000** as U.S. Application No. **09/600,621** or PCT International Application No. _____

and was amended on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is known to me to be material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Application No.	Country	Filing Date	Priority Claimed (Yes/No)
PCT/EP98/08480	PCT	12/28/98	Yes
98400104.0	EP	01/20/98	Yes

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States Provisional Application(s) listed below.

U.S. Provisional Application No.	U.S. Filing Date

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International Application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application or PCT International Application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is known to me to be material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application No.	Filing Date	Status (patented, pending or abandoned)

POWER OF ATTORNEY: I hereby appoint the following attorney(s) and/or agent(s) the power to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

Name: ANDREW G. GOLIAN	Registration No.: 25,293	
Send correspondence and direct telephone calls to: ANDREW G. GOLIAN	**E. I. du Pont de Nemours and Company** **Legal - Patents** **Wilmington, DE 19898, U.S.A.**	**Tel. No.** (302) 892-0747 **Fax No.** (302) 892-7343

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

INVENTOR(S)

Full Name of Inventor	Last Name: **ANDREIS**	First Name: **CHRISTINE**	Middle Name:	
	Signature (please sign full name):		Date: 8/9/00	
Residence & Citizenship	City: **MERZKIRCHEN**	State or Foreign Country: **GERMANY**	Country of Citizenship: **GERMANY**	
Post Office Address	Post Office Address: **HAUPTSTR. 2**	City: **MERZKIRCHEN**	State or Country: **GERMANY**	Zip Code: **D-54439**
Full Name of Inventor	Last Name: **FEYDER**	First Name: **GUSTY**	Middle Name:	
	Signature (please sign full name): Gusty Feyder		Date: 08/09/00	
Residence & Citizenship	City: **LUXEMBOURG**	State or Foreign Country: **LUXEMBOURG**	Country of Citizenship: **LUXEMBOURG**	
Post Office Address	Post Office Address: **159, RUE DES POMMIERS**	City: **LUXEMBOURG**	State or Country: **LUXEMBOUG**	Zip Code: **L-2343**

☐ Additional Inventors are being named on separately numbered sheets attached hereto.

Please type a plus sign (+) inside this box → [+]

PTO/SB/122 (11-96)
Approved for use through 6/30/99. OMB 0651-0035
Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

SEP 15 2000

CHANGE OF CORRESPONDENCE ADDRESS
Application

Address to:
Assistant Commissioner for Patents
Washington, D.C. 20231

Application Number	09/600,621
Filing Date	20 July 2000
First Named Inventor	Andreis
Group Art Unit	Unknown
Examiner Name	Unknown
Attorney Docket Number	2119-4203

Please change the Correspondence Address for the above-identified application to:

☐ Customer Number [ ] → *Type Customer Number here*

Place Customer Number Bar Code Label here

OR

☒ **Firm *or* Individual Name**	E. I du Pont de Nemours & Company				
Address	1007 Market Street				
Address					
City	Wilmington	**State**	DE	**ZIP**	19898
Country	USA				
Telephone	(302) 892-0747	**Fax**	(302) 773-0164		

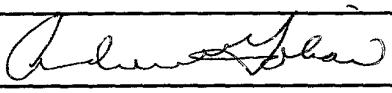
This form cannot be used to change the data associated with a Customer Number. To change the data associated with an existing Customer Number use "Request for Customer Number Data Change" (PTO/SB/124).

I am the :

☐ Applicant.

☐ Assignee of record of the entire interest.
Certificate under 37 CFR 3.73(b) is enclosed.

☒ Attorney or agent of record .

Typed or Printed Name	Andrew G. Golian, Registration Number 25,293
Signature	
Date	11 September 2000

Burden Hour Statement: This form is estimated to take 0.2 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.